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13. ABSTRACT (Maximum 200 words)

Nonlinear interactions of optical waves with droplets can be important in optical propagation through the atmosphere and in optical characterization of droplets. 1) We developed a detailed model of optical bistability in spheres which are illuminated with an intense optical plane wave. For the analysis we used the linear relation between the shift in the location of a morphology-dependent resonance (MDR) of the sphere and the change in the refractive index of an inhomogeneous sphere (when the change in index is real and small). 2) We applied the volume current method, a first-order perturbation solution for scattering, to inhomogeneous spheres. The scattering results obtained for a small off-axis sphere inside a sphere compare well with results computed using separation of variables. For a near-resonance sphere the results do not agree as well. 3) We modeled linear time-dependent internal fields in droplets illuminated with pulses that are Gaussian in space and time. 4) We completed the analysis of spheres which contain two-photon absorbers, and hence become inhomogeneous when illuminated with light. 5) We calculated scattering by droplets having the type of radially inhomogeneous refractive index that can occur in high-temperature combustion environments.

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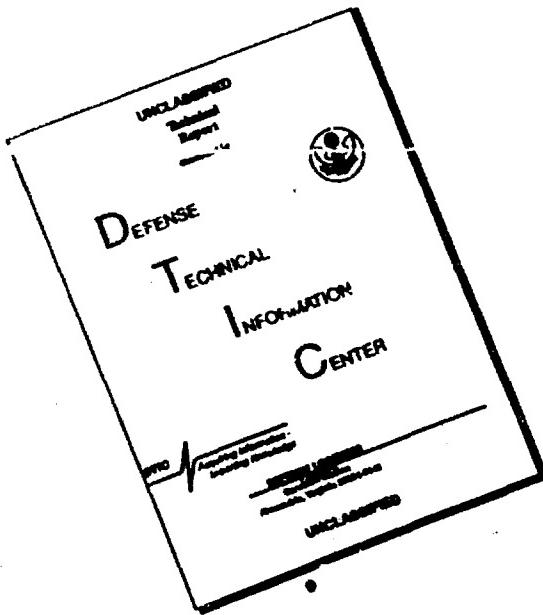
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COMPUTATION OF NONLINEAR OPTICAL SCATTERING BY DROPLETS

ARO Contract DAAL03-92-G-0308 to New Mexico State University

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1 THE PROBLEM STUDIED

Laser beams are being used to characterize the atmosphere. Lidar (Light Detection and Ranging) is increasing in importance. Lightwaves are also used for free-space communications. In using lightwaves to characterize or to modify the atmosphere, and in using lightwaves to communicate through the atmosphere, it is important to understand the interactions between optical radiation and the droplets or particles in the air. Linear and nonlinear optical scattering in droplets is also used to study growth and evaporation of droplets, adsorption of materials onto droplets, and chemical reactions in droplets. Light scattering is also being used to characterize droplets in sprays.

Laser beams used for characterization of the atmosphere often have intensities large enough to perturb the refractive indices of the droplets, i.e., to generate inhomogeneous droplets with refractive index (both real and imaginary part) distributions which are proportional to the internal intensity in the droplets. Consequently, the usual analyses of droplets which assume linear optical properties are not always valid, and methods for computing the nonlinear optical scattering by droplets are required.

Nonlinear optical effects such as phase modulation lineshape broadening, stimulated Raman scattering (SRS), stimulated Brillouin scattering (SBS), third harmonic generation (THG) and third-order sum frequency generation, and lasing have been observed in droplets. The effects typically occur in droplets with lower intensities than are required in bulk materials. The primary reason for the lower thresholds is the optical feedback provided by morphology dependent resonances, MDR's. If the refractive index of the droplet is a function of both time and space, because of its dependence on the internal intensity, the MDR's and quality factors, Q's, are also time varying.

Our objectives were to develop methods for computing the nonlinear optical scattering by droplets. Two classes of problems were considered: elastic scattering where the real part of the refractive index is intensity-dependent, and inelastic scattering where light is generated at new frequencies. An additional objective was to use the calculated results to help explain experimental measurements and to suggest new experiments.

2 SUMMARY OF MOST IMPORTANT RESULTS

We have developed techniques required for a more thorough analysis of nonlinear optics in droplets and have developed models for some nonlinear processes. We have continued to develop techniques for modeling the resonances of perturbed spheres, based primarily on perturbation methods. We have modeled linear time-dependent internal fields in droplets

illuminated with pulses that are Gaussian in space and time, to help us analyze the nonlinear optical effects usually observed with pulsed laser illumination. The main results are as follows.

1. Quality Factors and Effective Absorption and Gain Coefficients in Inhomogeneous Spheres

We developed an effective-average method for computing the scattering and absorption by slightly inhomogeneous spheres, and applied the method to the scattering by spheres which contain two-photon absorbers.

2. Light Scattering by Radially Inhomogeneous Droplets

We compared our separation of variables method for scattering by radially inhomogeneous spheres with a geometrical optics method developed by M. Schneider and Dan Hirleman.

3. Light Scattering by a Sphere Illuminated with a Gaussian Beam

We found that in a droplet the fraction of the energy in high Q MDRs increases as the illuminating beam is focused further outside the sphere. When a small-waist beam is focused outside the droplet, the majority of the internal energy tends to occur in the modes which have high Q's even when the spheres are lossy or illuminated with pulsed Gaussian beams.

4. Dispersive Bistability in a Dielectric Sphere

The shift in the resonance location of an MDR is linearly proportional to the maximum amplitude of the change in the refractive index of an inhomogeneous sphere if the change in index is real and small. The proportionality constant can be computed using the time-independent perturbation method. We have found that if the sphere is illuminated near an MDR so that one MDR dominates the field, then a simple expression for optical bistability can be written. When the sphere is illuminated further from a resonance, a much larger incident intensity would be required to generate bistable behaviour.

5. Volume Current Method for Scattering by Inhomogeneous Spheres

The volume current method, a first-order perturbation solution for scattering by inhomogeneous spheres, gives scattering results for a small off-axis sphere inside a sphere that compare well with results computed using separation of variables. However, for a near-resonance sphere the results do not agree as well. The method should be useful for computing nonlinear scattering by spheres.

3 PUBLICATIONS

3.1 Journal Articles Published or Submitted

E. E. M. Khaled, S. C. Hill, and P. W. Barber, "Scattered and internal intensity of a sphere illuminated with a Gaussian beam," IEEE Trans. Antennas Propagat., 41, 295-303, (1993).

D. Q. Chowdhury, M. M. Mazumder, and S. C. Hill, "Quality factors and effective absorption and gain coefficients in inhomogeneous spheres: application to two-photon absorption," submitted to IEEE J. Quantum Electronics, 29, 2553-2561, (1993).

E. E. M. Khaled, S. C. Hill, and P. W. Barber, "Light scattering by a coated sphere illuminated with a Gaussian beam," accepted in Appl. Opt.

E. E. M. Khaled, D. Q. Chowdhury, S. C. Hill and P. W. Barber, "Internal and scattered time-dependent intensity of a dielectric sphere illuminated with a Gaussian or a plane wave

pulse," accepted in *J. Opt. Soc. Am. A*.

E. E. M. Khaled, S. C. Hill, and P. W. Barber, "Internal electric energy in a spherical particle illuminated with a plane wave or off-axis Gaussian beam," accepted in *Appl. Opt.*

3.2 SPIE Proceedings Articles

S. C. Hill and R. K. Chang, "Model of nonlinear optical mixing processes in droplets," *Proceedings of the Conference on Laser Applications in Combustion and Combustion Diagnostics*, SPIE, 1862, pp. 309-321, (1993).

M. Schneider, E. D. Hirliman, H. I. Saleheen, D. Q. Chowdhury, and S. C. Hill, "Light Scattering by radially inhomogeneous fuel droplets in a high temperature environment," *Proceedings of the Conference on Laser Applications in Combustion and Combustion Diagnostics*, SPIE, 1862, pp. 269-286 (1993).

D. Q. Chowdhury and M. M. Mazumder, "Droplet diagnostics and scattering," *Proceedings of Conference on Laser Applications in Combustion and Combustion Diagnostics*, SPIE, 1862, pp. 164-181 (1993).

D. Q. Chowdhury, M. M. Mazumder, and S. C. Hill, "Linear and nonlinear scattering by a dielectric sphere," *Proceedings of Conference on Atmospheric Propagation and Remote Sensing*, SPIE, 1968, pp. 29-40 (1993).

3.3 Book Chapter

S. C. Hill and Richard K. Chang, "Nonlinear optics in droplets," *Proceedings of the 5th International Topsoe Summer School on Nonlinear Optics* (Aalborg, Denmark, 3-8, August, 1992) O. Keller, ed., Nova Science Publishers, New York, (*in press*).

3.4 Journal Articles Nearly Completed as of February 22, 1994

M. M. Mazumder, S. C. Hill, and D. Q. Chowdhury, "Dispersive bistability in a dielectric sphere."

S. C. Hill and H. I. Saleheen, "Light scattering by inhomogeneous spheres: approximate solutions using a volume current method."

4 PARTICIPATING SCIENTIFIC PERSONNEL AND DEGREES AWARDED DURING THIS REPORTING PERIOD

Steven C. Hill (PI)

Peter W. Barber. Now with Desert Research Institute, Reno, NV.

Dipakbin Q. Chowdhury, post-doctoral research associate, 80 percent support. Now with Corning Inc., Corning, NY.

Elsayed Khaled, received Ph.D, November, 1992. Partial support. Now an Assistant Professor of Electrical and Computer Engineering, Assiut University, Assiut, Egypt.

Mohiuddin Mazumder. Received support during the summer of 1992. Now is a Ph.D student at Yale University working with Professor Richard Chang.

Hasan Saleheen, received M.S. at New Mexico State University, November 1993. Received partial support. Now is a Ph.D. student at NMSU.

5 ACKNOWLEDGMENTS

Professor Richard K. Chang, Yale University, New Haven, CT suggested many of the problems we studied, and made many helpful comments on our work. The experimental results obtained by Professor Chang and his students Paul Chen, Ali Serpenguzel, Alfred Kwok, Christian J. Swindal, and Janice Cheng have provided a major impetus for the work done here. Dr. Ronald G. Pinnick (Army Research Laboratory, White Sands Missile Range, NM) also made many helpful suggestions, and provided a supportive environment in which to work.

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